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A REVIEW OF COUPLING AGENTS  
AS ADHESION PROMOTERS

By:

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and

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TRACOR, Inc.

2 September 1969

Prepared for National Aeronautics and Space  
Administration, George C. Marshall Space Flight  
Center under Contract NAS8-24073.



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## ABSTRACT

An extensive review has been made of the literature from 1950 to the present to include all work in the area of coupling agents and their use toward improving adhesion. The types of agents studied, mechanisms by which they act, substrates, adhesive systems and theories of adhesion are all considered.

By far the most common system is silane agent on a glass substrate, and a covalent siloxane linkage is proposed as the operative mechanism. Phosphorus esters and chromium-acid complexes are also known to be effective, and similar mechanisms are postulated. Other species will adsorb on adherend surfaces to provide an adhesive interlayer.

The use of coupling agents as admixtures with adhesives and the use and mechanisms of their behavior on metallic substrates has not been thoroughly studied until recently.





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## I. INTRODUCTION

The science of adhesion, although studied for years, is now experiencing serious investigation of the interfacial molecular forces responsible for this phenomenon. Discussions have arisen over the relative effects of chemical bonding and physical wetting, the best methods to obtain the optimum in each instance, and the role of adsorbed species and weak interfacial layers. It is not the purpose of this review to discuss the entire science of adhesion, but to concentrate on one area of increasing importance: the formation of a primary chemical bond linkage between a non-organic substrate and an organic adhesive system by the use of a coupling agent.

The fact that coupling agents are considered a principal factor in adhesion states a priori that the primary chemical bond is of considerable importance in adhesives. This is not as readily accepted as it may appear and is certainly not the whole story. Other matters which must be considered are wetting, interlayers, bulk properties, adsorbed species, environmental effects, etc. One should recognize that all of these parameters play important roles which cannot often be clearly separated.

Wetting of an adherend by the adhesive is, of course, necessary for good adhesion as shown many times over. After all, the function of the adhesive is to provide and maintain intimate contact between two solid surfaces. This is accomplished by having a liquid adhesive with a surface tension ( $\gamma_{LV}$ ) which is less than the critical surface tension of wetting ( $\gamma_C$ ) of the solid surface.<sup>346</sup> When this condition exists, the liquid forms a low (zero) contact angle with the substrate which denotes wetting and spreading on the surface. However, it is when one considers the micromechanics of the adhesive process that questions arise



as to the contribution from chemical bonding. It appears that primary chemical bonding to the substrate or the resin does indeed make a contribution although it is difficult to quantify.

Coupling agents may be used to form primary bonds to either or both the adherend and the bulk adhesive. The bond to the organic adhesive can occur by virtue of a reactive function on the agent similar to the monomer used or one which is reactive with the monomeric species. That is to say an agent may contain a terminal epoxy or vinyl moiety which will copolymerize with the organic system it contacts; or the agent may contain a function such as an amine which will cure an epoxy or urethane resin at least in part. Examples of each of these will be discussed below under the specific systems presented. However, with a polyester-glass system, the non-reactive silane, an amino alkyl silane, was shown to be detrimental to the physical properties. Evidently, it is possible for a release agent effect to occur if primary bonding does not exist between the agent and the organic phase.<sup>78a</sup>

Adhesion promoters which do not bond primarily to the resin phase are commonly long-chain fatty acids terminally substituted with *p*-chlorophenyl groups.<sup>225, 274, B-3</sup> It is postulated for these compounds that the carboxylic acid end groups are used to adsorb to surfaces of glass, metal and ceramic, while the pendant alkylphenyl groups offer a hydrophobic, resin-soluble phase.<sup>10, 53, 275, 144, 180</sup> These particular agents also increase the critical surface tension of the substrate to promote wetting.<sup>263</sup> However, in some instances, although the agent has the proper reactive moieties, it will decrease  $\gamma_C$  so that the adherend is wet only with difficulty.



There are, of course, two bonding mechanisms by which an agent can attach itself to a surface: simple Van der Waals attraction (dipole-dipole) or actual bond formation of a chemical bond by exchange of electrons. The first is evidenced by studies elucidating the relative adherence of substances with varying polarity of functional groups.<sup>303</sup> Studies with polymers containing acid and amide functional groups showed that both functions improved peel strength but that the former was superior.<sup>188</sup> It was concluded that electron availability contributes more than does dipole moment to interfacial interactions and therefore adhesion. This view corresponds to that which states that certain functions, such as piperidine, are chemisorbed to certain metallic surfaces, such as iron, to produce a relatively strong bond effective in adhesives, coatings or corrosion inhibitors.<sup>12</sup> This chemisorption phenomenon has been postulated to take place by virtue of the donation of the unbonded pair of electrons on the nitrogen to the unfilled 'd' orbital of the surface iron atom.<sup>12, 97b, 115b</sup>

Adsorption of simple polar long-chain compounds has been broken into three types: physical, chemisorption, and precipitated adsorption. The last two of which are very selective on metallic substrates. The mechanical properties and solubilities of each of these adsorbed films are distinctive.<sup>330</sup>

The formation of a direct primary bond, postulated for many coupling agents in the silane,<sup>290</sup> chromium complex,<sup>121</sup> and phosphate ester<sup>267</sup> classes, formerly was studied almost exclusively on glass substrates. Recently, however, such studies have been extended to metal surfaces.<sup>54b, 304</sup> The commercially available materials are compounds (M-X) which can be hydrolyzed to give reactive MOH functions which subsequently react with the SiOH surface functions by dehydration to form a very stable Si-O-M bond. These materials are generally used as a surface pretreatment



prior to bonding and are partially polymerized on the substrate before addition of adhesive. An entirely new surface then presents itself to the adhesive, a surface which is wettable and reactive with the polymer.

Epoxy resins form their own primary bonds to some extent. Free radicals which form during the cure produce chemical bonds with metallic adherends.<sup>21, 22</sup> In the realm of attaching adsorbable functions to the polymer backbone some limited success has been seen.<sup>102b, 253</sup> Where one has the bondable functions on the backbone of an applied polymer new considerations are necessary. One is that in order for the groups to be effective and reach the substrate surface, the conformational energy of the polymer chain must be overcome. That is, the adsorbable functions must become aligned and in doing so possibly distort the normal chain equilibrium conformation.

However, when a polymeric rather than monomeric adsorber is utilized, significant increases in the adhesion of monomolecular layers is realized. This was dramatically shown for corrosion inhibitors of various molecular weights where a degree of polymerization greater than 4 (adsorbable units) gives an increase in corrosion inhibition by a factor of 10,000.<sup>12, 97b, 115b</sup> The cause of this synergistic effect has been discussed in terms of the equilibrium between each chemisorbed group and the surface. With a multitude of bonded moieties on a polymer backbone, those few which are temporarily non-bonded in equilibrium cause essentially no disruption in overall attachment of the polymer.<sup>343</sup>

Recent work has elucidated the role of the resin in the vicinity of the interface, where, through coupling agents, a modified region of adhesive is produced.<sup>157</sup> With no agent and poor adhesion, debonding and failure occur at the interface.



However, with covalently bonded agents, failure is essentially cohesive deep in the resin body rather than adhesive at the substrate. (Good adhesive systems are known to fail cohesively.) Further improvement is realized when an additional "inner layer" or "interphase" is introduced.<sup>157, 198</sup> This intermediate layer has been used on a macro-scale by mixing the adhesive and agent in high concentration and applying in a thin coat as a prebonded primer.<sup>202</sup> It is postulated through mathematical stress and modulus analyses that a thicker interphase will enhance yield stress. This region of modified resin is an extension of that produced by a monomolecular layer of the coupling agent. This serves to decrease the stress concentrations over a larger distance and to increase bond strength between two drastically different materials.

It is also possible, however, to effect an apparent increase in bond strength without changing interfacial forces. If one plasticizes the bulk adhesive an increase in strength is seen. This is due only to absorption of energy by the organic matrix.

The use in composites is an indication of what is seen in filled polymeric systems. As would be expected, as one increases the interaction between filler and organic matrix, the effect of the filler is magnified.<sup>217</sup> Stated another way, the ability of the fillers to enhance matrix properties is limited to and dependent upon the state of bonding at the polymer--filler interface.<sup>25</sup> Fillers show an effect similar to that of increasing the crosslinks in a system, e.g., an increase of glass transition temperature. This is due to the fact that polymer-surface interactions tend to limit the mobility of the polymer molecule much as does crosslinking.<sup>180, 181, 302</sup> Coupling agents then decrease the molecular mobility at the surface even more effectively than simple surfaces and essentially do produce a crosslink at the



surface. This was shown by impregnating glass fibers with  $\text{TiCl}_4$  and placing these in an olefinic monomer, styrene, resulting in initiation of polymerization at the glass surface. The formation of a chain coagulation structure of particles when properly treated<sup>306</sup> is possible. Thus, a polyvinylchloride filled with  $\text{TiO}_2$  which was modified with octadecylamine demonstrated this ultimate effect of fillers.

Aside from initial bond strength improvement there have been two other distinct advantages generally recognized as being due to coupling agents. These are: (1) a lesser dependence on cure schedule for optimum strength, and (2) a resistance to bond strength degradation by aging or moisture. The second and perhaps more serious effect has been borne out experimentally on glass reinforced thermoset and thermoplastic systems<sup>76, 229, 308</sup> and with metal-polyurethane bonds.<sup>304</sup> This phenomenon is attributed by DeLollis to the fact that water is preferentially adsorbed on the substrate with subsequent displacement of any adhesive which is not primarily bonded. However, when a coupling agent is utilized, a primary bond is formed to the adherend surface which is not susceptible to hydrolysis or displacement by water. Thompson and Hill<sup>304</sup> have also noted that only when a silane adhesion promoter is used in a urethane adhesive can the original strength of a water saturated weakened bond be regained by drying the sample.

The first effect, dependence of bond strength on cure schedule, was elucidated when lap-shear strength was determined for an epoxy-aluminum system with varying cure schedule.<sup>54c</sup> This strong dependence of adhesion on cure schedule was decreased considerably by the use of coupling agents. This means that manufacturing procedures can be less strictly controlled.



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Certain generalities are worthy of note from this review. (1) Most of the work on coupling agents, both from a practical and theoretical standpoint, has been done on glass substrates. Recently, however, there has been increasing interest in the effect of coupling agents on metal surfaces.<sup>54b, 304</sup> (2) Most experiments have been done by pretreating the surface with the coupling agent prior to application of the adhesive. Again, interest is developing in the incorporation of the agent into the adhesive system because of the economic advantage of a single application.<sup>54b, 304</sup> However, the integral mixture requires not only that the agent migrate to and react at the substrate surface, but also that functional groups be similar in reactivity to the resin so that polymerization is realized only when desired.

The following sections discuss in detail all coupling agents known through June, 1969. The three principal types are silanes, chromium complexes and phosphorus esters which are discussed separately. A final section is a review of many miscellaneous surface agents on which some experiments have been run. A table of comparative data is also given.

The bibliography has been selected from over 1100 references on this subject since 1950. Its alphabetical listing by authors is supplemented by location of the abstract in Chemical Abstracts. Furthermore, following each reference is a series of numbers indicating subject matter dealt with therein according to the following code. A second portion of the bibliography, Part B, is a compilation of books and review articles on coupling agents and adhesive technology.



6500 TRACOR LANE, AUSTIN, TEXAS 78721

I. Adhesive

1. Epoxy
2. Urethane
3. Elastomer
4. Polyester
5. Phenolic
6. Silicone
7. Other

II. Substrate

11. Copper
12. Aluminum
13. Steel
14. Glass
15. Rubber
16. Plastic
17. Other

III. Agent Type

21. Phosphorus
22. Chromium
23. Piperidine
24. Silane
25. Other

IV. Miscellaneous

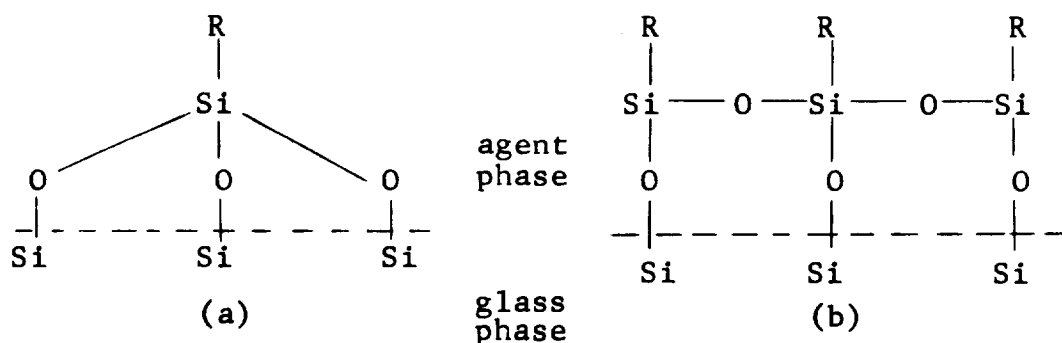
31. Environmental Effects
32. Surface Chemistry
33. Surface Preparation
34. Fillers
35. Test Methods



## II. SILANES

A large number of silanes of general structure  $R_{(4-y)}SiX_y$  have been evaluated as coupling agents. In these compounds the R groups are organofunctional groups which can be chosen for a specific purpose. The X groups may be halides, alkoxides, and/or acyloxy groups -- all of which are hydrolyzed under the conditions of application to give  $Si(OH)_y$  groups. These silanol groups may in turn react with polar surface groups of the substrate by dehydration to form a primary bond. Thus, the molecule is ambifunctional; it contains polar silanol groups capable of adhering to the surface of glass, metals, etc., and a group R tailored to interact with the adhesive resin.

The adherence of the polar hydroxyl groups to the surface of glass is postulated to be through an ether type (siloxane) linkage between the  $SiOH$  groups of the glass and the  $Si(OH)_y$  groups of the silane. Each molecule has the possibility of forming  $y$   $Si-O-Si$  bonds. Whether the siloxane bonds are all formed



with the glass surface (as in a) or with adjoining silane coupling agents (as in b) is still in question. However, it is known that the greater the possible number of siloxane bonds, the greater the stability of the linkage between coupling agents and the surface.<sup>343</sup> Further, kinetic studies have shown an increase in bond order between silane and glass in the order of mono-, di- and trichlorosilane.<sup>117</sup> Consequently, hydrolysis is

more difficult and water stability is greatly enhanced for glass-resin bonds using as coupling agents silanes of the type  $R-SiX_3$ . While many silanes have been tested<sup>19, 156, 336, 326</sup> it is logical that the most effective and commercially available coupling agents have the general structure,  $RSiX_3$ .

Considerable work has been done in an attempt to determine whether the interaction between substrate and silane is actually a chemical bond as postulated above or simply a physical adsorption. Thermograms of the treatment of quartz powder with  $CH_3SiCl_3$  vapor show a pronounced exothermic effect which is explained in terms of bond formation. Silanes with fewer  $X$  groups,  $(CH_3)_2SiCl_2$  and  $(CH_3)_3SiCl$ , show proportionately weaker bonds.<sup>11</sup> These results were further substantiated by infrared spectroscopic studies of deuterated glass surfaces which showed weaker bonding on mono- and difunctional silanes as compared to the trifunctional silanes. Only external hydroxyl groups of the glass were found to react and these on a 1:1 basis with  $(CH_3)_2SiCl_2$  and  $(CH_3)_3SiCl$ . The  $CH_3SiCl_3$  reacted more on a 1:2 and 1:3 basis with the glass hydroxyl groups.<sup>52, 59, 102, 117</sup> Isotopically labeled ( $^{14}C$ ) silanes have also been used to examine the extent of reaction with glass surfaces. Tracer and photomicrographic studies indicate continuous films instead of islands of the coupling agent with covalent bonds at the surface. Failure of the film in boiling water was attributed to failure of the glass substrate and not the glass-silane interface.<sup>145, 325</sup>

A novel investigation of the electronic interaction between coupling agent and glass substrate involved the determination of charge developed by stripping films of polymers from the glass surface. With untreated glass, the charge on the stripped film was negative, but it was positive when the glass had been treated with silane.<sup>167, 168</sup> The study is indicative of greater electronic



interaction than can be explained by simple absorption of the coupling agent. Chemical methods have also been used with some success to study glass-silane interaction.<sup>19, 171, 337</sup>

Although silanes are useful as coupling agents on substrates other than glass (aluminum, steel, copper, etc.)<sup>54b, 304</sup> no definitive work has been published on the mechanism of interaction between the silane and these surfaces. Proposed mechanisms involve either a silanol-type interaction with surface oxide or hydration layers, or chelation of the metal by aminoalkyl groups of the silane.

Silane coupling agents can be divided into three classes according to their reactivity with the organic phase, non-reactive reactive, and catalytic, and are discussed below accordingly.

Non-reactive silanes have R groups which are alkyl or aryl with no unsaturated or other reactive moieties. With a short alkyl group as in  $\text{CH}_3\text{SiCl}_3$ , a finish which is non-adherent to polar substances such as water and polyurethanes can be imparted to surfaces.<sup>19, 80, 94, 102a, 103, 104, 163</sup> However, methyl silanes have been used with excellent results as a primer for silicone rubber bonding to many surfaces.<sup>120</sup> Usually longer alkyl groups,<sup>84, 278</sup> aryl groups,<sup>78a, 32, 226, 332</sup> aralkyl groups,<sup>221, 225</sup> and halogenated derivatives of these groups<sup>122, 225, 259</sup> are incorporated in the silane for cohesive interaction with the bulk adhesive.<sup>116, 197, 239, 278</sup> Such groups are effectively used with non-polar resins such as polystyrene and other poly-vinyl compounds.

Reactive silanes contain a function capable of chemically reacting with the bulk adhesive commonly as a comonomer. Reactive functions commonly employed are vinyl groups,<sup>9, 61, 111, 114</sup> epoxides,<sup>31, 33, 34</sup> mercaptans,<sup>105</sup> alcohols,<sup>106, 175</sup> phenols,<sup>197</sup> acids,<sup>60</sup> esters,<sup>82, 85</sup> and combinations of these functions.<sup>153, 281, 295</sup> The choice of reactive group depends on the adhesive to be



used and the type of interaction possible. Obviously, compatibility of the R groups and the resin is necessary even in the case of the non-reactive silanes. To provide compatibility, polarity of the resin should match the polarity of the functional group to the extent of matching the degree of hydrogen bonding in each. A summary of the various reactions which may occur between resins and functional groups has been prepared by Plueddemann, et.al.:<sup>242</sup>

- (1) Condensation reactions are possible between hydroxyl, carboxyl, mercapto, amino, or epoxy groups on silicon with hydroxyl or carboxyl groups of a polyester.
- (2) Olefinic hydrocarbons or unsaturated ether and ester groups on silicon may participate in free-radical-induced addition polymerization with an unsaturated polyester and styrene monomer.
- (3) Almost all functional groups are capable of reacting with an epoxy resin or its curing agent in an epoxy laminate.

The third class, the catalytic silanes, contain amino groups which can act as catalysts in the polymerization of phenolic, urea, and melamine resins and can act as curing agents for epoxy<sup>17,160, 183, 294, 296, 313</sup> and polyurethane<sup>6</sup> resins. Thus, the coupling agent is integrally involved with and bonded to the bulk adhesive. Examples of the amino groups present in commercially available catalytic silanes are  $-(CH_2)_3-NH_2$ ,  $-(CH_2)_3-NH-(CH_2)_3-NH_2$ , and  $-(CH_2)_3-NH-(CH_2)_2-NH-(CH_2)_2-CO_2CH_3$ .<sup>9, 15, 85, 102a, 118</sup> In reaction with some adhesive systems, the amino silanes may be classified as simply reactive silanes and not catalytic. As such the amino group is capable of reacting with alkylhalides, acids, anhydrides, and esters in substitution or condensation reactions. (Oddly, the amino silanes give poor adhesion to polyester resins, perhaps because of amine inhibition of the polyester cure.)<sup>242</sup>



Silane coupling agents are usually applied to a glass substrate surface as a pretreatment from a dilute aqueous solution. Acid or base is added to the solution to aid in the hydrolysis of the  $\text{SiX}$  groups to  $\text{SiOH}$ . The solution is necessarily dilute (0.5-1.0% coupling agent) since optimum coupling effect is achieved with a mono-molecular layer rather than a multi-layer film of the silane. The surface is then dried, usually at temperatures over  $100^\circ\text{C}$ , to promote the condensation reactions and to remove excess water. Vapor phase application of alkyl-alkoxy silanes is accomplished by mixing the silane with moist air for hydrolysis and applying it to nascent glass surfaces while the temperature is held near the decomposition point of the silane.<sup>89</sup>

Some integral blending of the coupling agent into the adhesive resin has been tested with encouraging results.<sup>6, 27, 36, 54b, 87, 97a, 105, 108, 115a, 123, 158, 234, 290, 292, 304</sup> For example, outstanding improvement of metal bonding has been reported when silane coupling agents were integrally mixed with urethane and epoxy adhesives.<sup>54b, 304</sup> Such application has the economic advantage of reducing the number of steps required in the adhesive process. However, this advantage is somewhat negated in the usual procedure for integrally mixed adhesive systems because (1) a slower curing process may be needed to allow migration of the coupling agent to the substrate surface and (2) an excess of coupling agent is usually required since apparently all of the coupling agent does not reach the adherent surface.

The preceding discussion has dealt primarily with silane glass surface systems. More work has been published in this area than about other substrates because of the long-time interest in glass lamination in composites. However, silane coupling agents, as surface pretreatments or as mixtures, are being used increasingly in the bonding of metal substrates with a variety of

adhesives.<sup>54b, 304</sup> The brief discussion below of some of the systems which have been reported will give some indication of the extent of such usage.

First, it should be noted that improved properties other than total strength may be obtained through the use of silane coupling agents with certain adhesive systems. The results of a long-term aging study have shown that more reliable and much improved adhesive properties were achieved when silane coupling agents were used with polyether-based polyurethane adhesive on aluminum. During a three-year period, loss of strength with normal variations in atmospheric humidity has been minimized, and greatly increased strengths at room temperature and +200°F have been observed.<sup>304b</sup> Furthermore, the samples utilizing a coupling agent can be dried to reproduce the original strengths.

Copper, treated with aminoalkyl silane compounds, was effectively bonded to silicone elastomers with exceptional bond stability at elevated temperatures.<sup>34, 234a</sup> Considering the substrate and adhesive, the probable mechanism was the chelation of the copper with the amino groups.

Aluminum has been treated with silanes to improve bonding with several adhesives. Bonding to silicone elastomers was enhanced either by amino silanes incorporated into the silicone<sup>49, 70, 182, 248, 249</sup> or by methyl silane treatment of the metal surface.<sup>97, 120</sup> Vinyl silane treatment of the metal also aided the adhesion of silicone elastomers.<sup>138, 156</sup> Adhesion of polyvinylhalides to aluminum and steel surfaces was increased by addition of amino silanes.<sup>277</sup> Polyurethane bonding of aluminum was greatly increased (particularly at +200°F) by surface treatment of the metal with an amino or epoxy containing silane.<sup>304</sup> Furthermore, incorporation



of amino and epoxy silanes into the polyurethane adhesive has been tested on aluminum with good results.<sup>54b, 87, 260, 304a,b</sup> Amino and mercapto silanes have been proved effective as integral parts of adhesive mixtures of epoxy,<sup>54b</sup> phenolic, and polyvinyl butyral resins.<sup>244</sup>

Steel plates bonded with an epoxy-silane-modified polyurethane adhesive withstood 24 hours in boiling water as compared to only one hour with the unmodified adhesive.<sup>6</sup> Similarly, the incorporation of an epoxy or amino silane into an epoxy resin increased the sea-water resistance of the resin coating on steel.<sup>30, 86</sup> This serves to point out the contention that the primary benefit of coupling agents may be their effect on aging characteristics rather than on total initial strength of the adhesive.<sup>76, 304</sup> The proper choice of coupling agents will also allow effective bonding of "contaminated" steel. An epoxy silane treatment of oily or rusty steel increases the lap-shear strength of epoxy bonds over those of clean metal or untreated contaminated metals.<sup>85</sup>

Bonding of rubber to metal substrates such as copper, aluminum, or steel was improved by the use of amino silanes either as a pretreatment of the surface or by incorporation into the rubber.<sup>182, 248</sup> Similarly, amino-silane treated glass showed better adhesion to synthetic rubber such as neoprene.<sup>191</sup> In addition, a "double" treatment of glass with (1) an unsaturated silane followed by (2) an elastomer containing a free-radical curing agent was shown to increase the adhesion to natural or synthetic rubber.<sup>317</sup>

Use of coupling agents in bonding of plastic materials usually has been limited to lamination procedures in which the silane was applied to the reinforcing material and the organo-functional group interacted with the resin as it polymerized.<sup>191, 210, 258, 282, 316, 319, 323</sup> A study of the structural features of silanes which increase wet and dry flex strength of glass-polybenzimidazole



laminate at high temperature (600°F) has concluded that two conditions are necessary: (1) a stable aromatic bond to silicon and (2) a reactive functional group for bonding to the resin. Thus, cyanophenyl, carboxyphenyl, and bromotolyl groups were adjudged as most effective.<sup>238</sup>

Surface treatment of polymeric adherends with coupling agents has been done.<sup>124</sup> Impregnation of polyester and polyamide fabrics with glycidyl silanes has been used as a pretreatment for coating the fabrics with silicone rubber prior to polymerization and vulcanization.<sup>81</sup> Amino silanes have been used to treat a polyester film used for heat-pressure bonding to polyethylene film to give a highly water-resistant bond.<sup>83</sup>

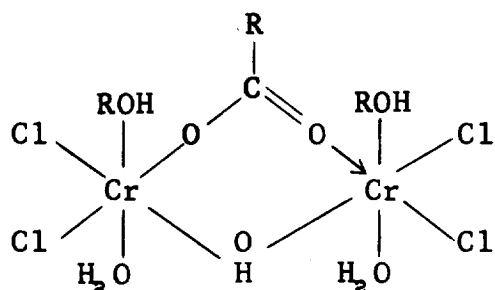
Some substrates to which silanes have been applied must be termed "miscellaneous." A priming of clay surfaces by amino silanes is claimed to increase the adhesion of polyurethane and epoxy coatings and to aid in the water resistance of these coatings.<sup>287</sup> Several adhesive mixtures have been formulated which contain various silanes as integral constituents. These adhesives have been effectively applied to a variety of substrates: paper, brass, wood, ceramics, etc.<sup>57, 271</sup>

Much of the preceding discussion has centered on the use of silanes with the widely used adhesives--the epoxides, urethanes, polyesters, etc. The development of new adhesive resins such as polyimides, ionomers, and polysulfones will require the development and evaluation of more silane coupling agents tailored for each of these resins.

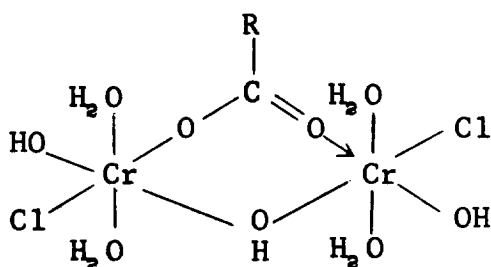


### III. CHROMIUM COMPLEXES

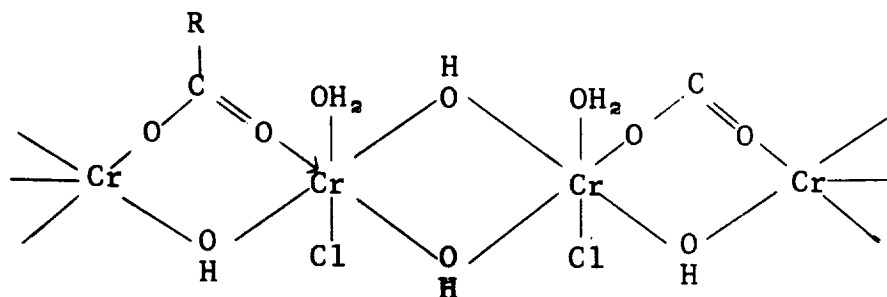
Trivalent chromium complexes which include an organic acid anion as one of the complexing species are widely used as coupling agents. The charge on the complex is determined by the coordinating groups which may be anions (usually chloride ions) or neutral molecules (water or alcohols). The structure of such a complex may be pictured as:



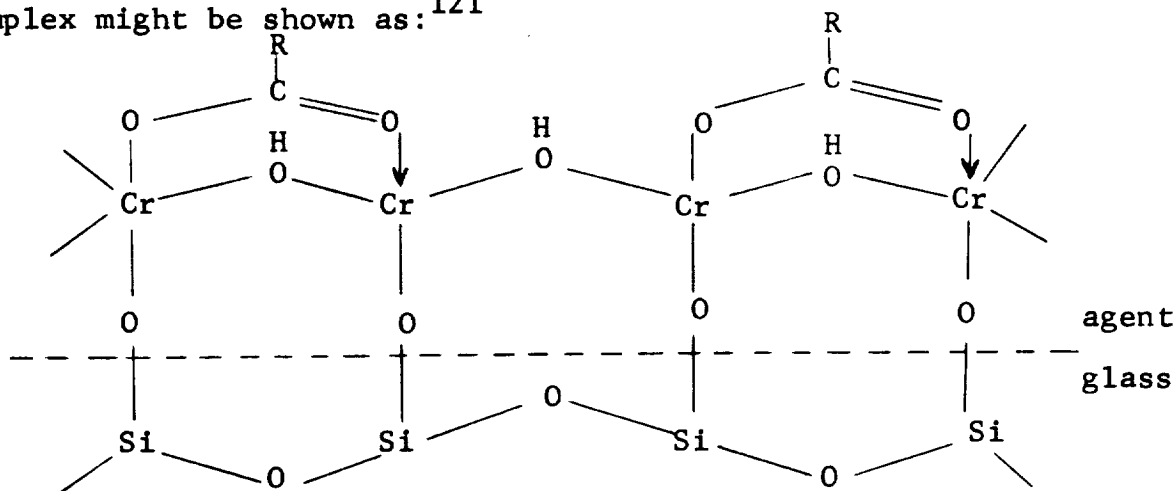
Dilution with water, particularly within pH ranges from 5 to 7, begins hydrolysis in which some chloride ions are replaced by hydroxyl groups as shown below:<sup>343</sup>



As hydrolysis continues, polymerization begins by the process of olation whereby the complexes are connected by hydroxyl groups:



Olation continues with a resulting increase in the size and positive charge of the complex. Absorption of the complex by a substrate surface may occur at any time in the olation process and is thought to occur at negatively charged sites on the surface. Thus, acidic positions such as SiOH in glass will attract the complex if the pH is high enough to encourage ionization to  $\text{SiO}^-$ . The pH necessary for effective coupling varies with the type of glass due to variation in ionization of acid groups with glass composition. The bond between a glass surface and the complex might be shown as:<sup>121</sup>



Interaction of the chromium complex with the adhesive is of three types:<sup>343</sup>



- (a) Coordinate covalent bonding between carboxyl, alcohol, and amine end groups.
- (b) Reactions between Cr-Cl and CrOH groups with alcohol end groups to form Cr-O-R linkages with the elimination of either HCl or H<sub>2</sub>O.
- (c) Copolymerization between the double bonds of methacrylic acid and reactive vinyl type double bonds in the resin.

Interaction of types (a) and (c) depend largely on the type of acid used in making the complex. The acids can be broadly classified as having reactive or non-reactive R groups. Reactive R groups are unsaturated (for copolymerization) or contain functional groups capable of participating in hydrogen bonding. In addition to methacrylic, acrylic,<sup>337</sup> phthalic,<sup>3</sup> and succinic acids<sup>230</sup> have been used as "reactive" complexing acids.

"Nonreactive" acids are exemplified by stearic acid which is used principally to impart a hydrophobic finish to surfaces<sup>166</sup> and does not aid adhesion of glass to polyester resins.<sup>121</sup> However, stearic acid-chromium complexes are useful in treating CaCO<sub>3</sub> filler for such resins. Complexes of several carboxylic acids, saturated and unsaturated, mono- and dibasic, were evaluated for effectiveness in the surface treatment of CaCO<sub>3</sub> filler for absorption of polyester resins, and caproic acid was found to be superior.<sup>2</sup>

Chromium complexes have been used mainly to treat glass surfaces with only limited application to other substrates. Application is made by dipping the glass surface in a dilute aqueous solution of the acid-chromic chloride complex whose pH is maintained near 6. Aging of the complex to allow olation and polymerization has been shown to greatly improve performance as a coupling agent. The surface is then dried at ~150°C prior to the addition of the adhesive polymer. (An exception in which

no adhesive is used is a novel method of preparing inner-reflecting glass spheres by dropping hot spheres into a cold solution of methacrylato-chromic chloride. The complex is the bonding agent which holds the fractured spheres together.<sup>339)</sup>

Other substrates to which chromium complexes have been applied include cellulose,<sup>100</sup> mineral fibers,<sup>78b</sup> rubber,<sup>69</sup> leather and textiles.<sup>3, 166</sup> Cellulose and mineral fibers were treated to increase their adherence to polyolefin films. Rubber was given increased moisture resistance by treatment prior to vulcanization. Hydrophobic finishes were imparted to leather, paper, and other textiles by treatment with a stearic or oleic acid complex. A related process involves addition of chromium salts to polyvinyl alcohol and polyacrylic acid to increase their elasticity, water resistance and film-forming capacity.<sup>230</sup>

Chromium complexes have been used as adhesion promoters with a variety of adhesives. Epoxy resins have been widely used in joining treated glass surfaces,<sup>109, 131, 227</sup> in bonding treated mica in electrical insulators,<sup>149</sup> and in moldings with treated SiO<sub>2</sub> filler.<sup>252</sup>

Polyester adhesives have been used to bond treated glass surfaces.<sup>1, 126, 129, 216, 275, 334</sup> Chromium-complex-treated sand filler has been molded with polyester resin to produce stain-resistant cast-resin surfaces.<sup>51</sup> Combinations of polyester and epoxy resins have been used on treated glass laminates<sup>8</sup> and in molding treated mica filler.<sup>149</sup>

Phenolic resins have been used to laminate glass fibers which had been previously treated with methacrylato-chromic chloride.<sup>44, 129</sup> Phenolic, epoxy, polyester, and organo silicon resins were tested alone and in combinations for adhesion to treated glass surfaces<sup>109</sup> where epoxy resins were found to be superior.



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Several investigations<sup>27, 126, 129, 131, 275</sup> have compared the efficiency of chromium complexes and silanes in promoting adhesion of glass substrates to various adhesives. The silanes were always found to be superior particularly with polyester adhesives.

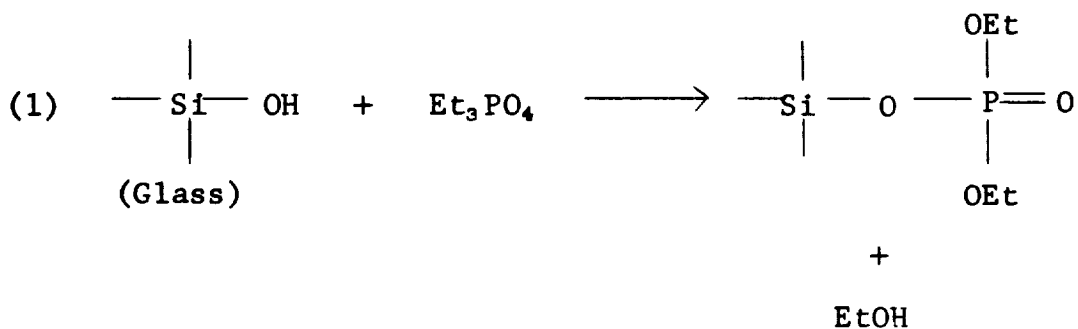
While chromium complex coupling agents increase dry bond strengths significantly, their real value is in increasing wet strength or moisture resistance of bonds. For instance, dry strength of glass-epoxy laminate was increased by 30% while wet strength was increased by 100% over that of the untreated substrate.<sup>67</sup> However, in one case<sup>27</sup> methacrylic acid-chromium complex treated glass was found to have poorer resistance to water absorption than untreated glass when bonded with polyester. These poor results are attributed to experimental and handling techniques, since improvement in wet strength is obtained if the Cl/Cr mole ratio is between 0.1 - 1.2 (optimum 0.6 - 0.7) when the methacrylate-chromium complex is prepared. The Cl/Cr ratio is adjusted by varying the amount of  $\text{CrO}_3$  and HCl added to methacrylic acid.<sup>307</sup>

#### IV. PHOSPHORUS CONTAINING COMPOUNDS

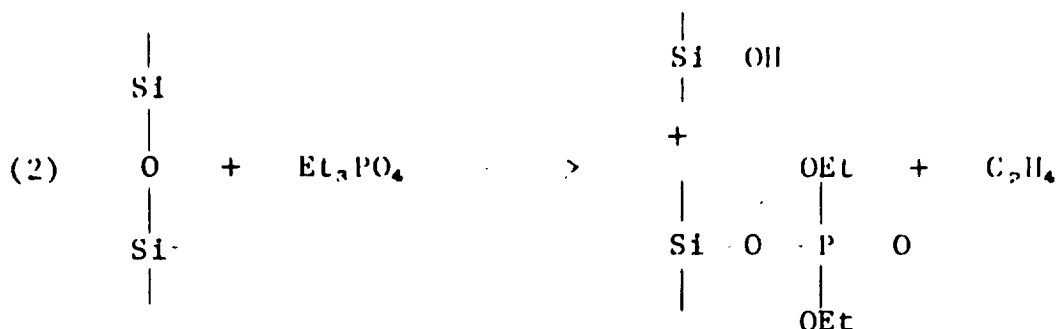
Some phosphorus compounds are known to behave as coupling agents. Esters of phosphoric, phosphonic, and phosphorous acids react with epoxy resins and decrease their viscosities.<sup>133</sup>

Wollastonite, when treated with  $H_3PO_3$  before addition to a polyester containing tritolylyl phosphate (TTP), contributes significantly to the flexural strength of the filled material.<sup>206</sup> In place of TTP one can utilize phosphate esters substituted with allylic, vinyl, ester, and amino alkyl groups. It is postulated that the mineral filler is bonded to the polymeric matrix by virtue of a phosphorus linkage.

A considerable amount of definitive work has been done with these phosphorus agents at the Naval Applied Science Laboratory by Schrader, Lerner, D'Oria and Deutsch.<sup>263, 264, 265, 266, 267, 268</sup> Triethylphosphate was found to adsorb irreversibly on E glass surfaces in a high vacuum system. Desorption/decomposition experiments at high temperatures indicated the original adsorption mechanisms to be:



and



Such treatments once again showed improved flexural strengths when used on E glass cloth in epoxy laminates.<sup>266</sup>

Next, a study with TTP labeled with <sup>32</sup>P and <sup>14</sup>C showed two reactions to occur at the surface: (1) the formation of a primary chemical bond, Si-O-P, between TTP and the glass surface; and (2) hydrolysis of TTP by adsorbed water to give cresol and H<sub>3</sub>PO<sub>4</sub>,<sup>265</sup> which contributes to De Lollis' desorption mechanism.<sup>76a</sup> Further tracer studies showed a three-component film present upon adsorption of diethyl phosphite (DEP): (1) a water soluble material at 25°C which is physically adsorbed, (2) a species chemisorbed through a single Si-O-P bond which is insoluble in water at 25°C, and (3) a water insoluble portion at 100°C which is bonded through multiple Si-O-P bonds. The physically adsorbed layer is believed to participate in increasing the dry strength of epoxy laminates.<sup>264</sup> The general method for applying DEP to glass is to place the substrate in a 1% aqueous solution of DEP, air dry, and heat at 150°C for 10 minutes.<sup>263</sup>

Similar multi-component films were found with <sup>14</sup>C labeled (γ-aminopropyl)-triethoxysilane (A-1100).<sup>267</sup> An increase in the temperature of adsorption of these agents increases the stability (irreversibility) of the layer. This effect is due to the fact



that at higher temperature, a polymeric monolayer is formed with multiple sites of attachment to the surface.<sup>268</sup> Thus, when one surface bond is reversibly broken by hydrolysis it has time to reform before other functions are hydrolyzed. This is the same synergistic effect experienced by Hackerman and coworkers<sup>11, 97b, 115b</sup> when studying polymeric piperidinyI corrosion inhibitors. They noticed an increase in corrosion inhibition (by chemisorption) by four orders of magnitude when the degree of polymerization of the adsorbed species was greater than four.

Organophosphorus compounds have also shown utility in increasing the adhesion of polymeric substrates to metals and other organic adhesive systems. Thus, radiation grafting of  $(ClCH_2CH_2O)_2 P(O)CH:CH_2$  to polypropylene improves its adhesion to metals with only a small change in bulk properties.<sup>179</sup> This method also allows the study of interfacial effects independent of the bulk properties. This same study claims that interfacial effects act only up to a critical minimum value above which the bulk mechanical properties of the adhesive are the sole determinants of total strength. This contributes to the observation of the high strengths obtained when one can achieve cohesive failure.

Similarly, ethylene-vinyl acetate copolymers show increased adhesion to metals when they are peroxide crosslinked in the presence of trialkylphosphate, where the alkyl groups are incorporated into the backbone leaving pendant phosphate moieties.<sup>67</sup>

Thermoplastic solution adhesives containing TTP are used for bonding PVC to aluminum, steel to zinc, and for ABS to aluminum.<sup>5</sup>

Even fluorinated polyolefins show adhesive improvements on treatment with phosphites. Dipping poly(trifluorochloroethylene) in dibutyl phosphite at 150°C allows it to be bonded to an acrylate-acrylic acid copolymer sheet under pressure.<sup>68</sup> The mechanism in





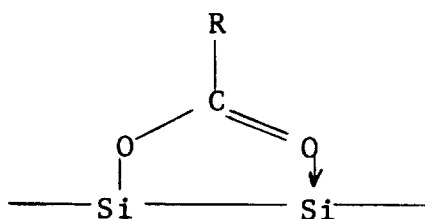
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this case may be different than with metallic substrates in that here a transesterification of the surface phosphite may be the bonding mode.

## V. MISCELLANEOUS AGENTS

### A. Acids

Several different types of carboxylic acids have been used as coupling agents on metals, glass, and other substrates. The polar carboxyl group has the capability of coordinating or reacting with polar surface groups such as the SiOH of glass or the AlOH of aluminum in much the same fashion as do chromium complexes or phosphorous compounds:



Probably the stability of this complex is reduced because of the fewer coordinating positions of the carboxyl group relative to the chromium or phosphorous compounds. The presence of an ortho-hydroxyl or thiol group in the more effective salicylic<sup>137, 177</sup> and thiosalicylic acids<sup>231</sup> offers the possibility of a third coordinating position which would increase stability. Salicylic acid has been incorporated into a phenolic resin to form an adhesive which is effective on a wide variety of substrates.<sup>177</sup> Salicylic acid has also been used as a pretreatment of glass for resin adherence<sup>137</sup> and thio-salicylic acid as a pretreatment of cord fabric to increase rubber adhesion.<sup>231</sup>

The carbon chain, R, has been varied considerably in the evaluation of coupling effectiveness of acids. The same concepts apply to the R groups of the acids as to those of the silanes and chromium complexes, and the group may be reactive or nonreactive toward the adhesive resin. The seventeen-carbon saturated R-group



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of stearic acid exemplifies the nonreactive type. It has been shown to increase the bond strength of metals bonded with polyvinyl-formal-phenol resin by 5-500% if applied as a 0.02-0.06% part of the resin, whereas higher concentrations weakened the bond.<sup>215</sup>

Other investigators have shown the value of a mono-molecular layer of the acid applied to the substrate. Application has been made either by carefully dipping the substrate in water which is covered by a monolayer of stearic acid<sup>10</sup> or by electrophoresis.<sup>255</sup> Stearic acid has been shown to greatly improve the moisture resistance of polyethylene bonds between treated aluminum plates.<sup>10, 62, 324a</sup>

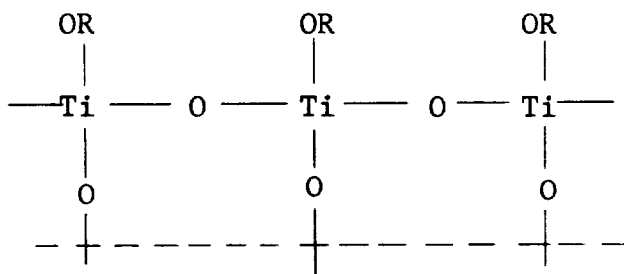
The extraordinary effectiveness of the *p*-chlorophenyl group as the terminal substituent of *R* groups in increasing adhesion is attributed to the relatively high-energy chlorophenyl outer surface exposed to the adhesive resin, thus producing a surface which is easily wet by the adhesive. The *p*-chlorophenyl moiety has been used as a terminal substituent on 12, 14, 18 and 20-carbon carboxylic acids and as an ethyl silane substituent.<sup>18, 225, 273</sup>

Acids with reactive *R* groups, such as maleimidopropionic or succinimidopropionic, have been used as coupling agents on several metals and alloys for epoxy bonding. Aluminum bonded in this manner showed good resistance to boiling water for 48 hours.<sup>274</sup> Incorporation of unsaturated acids into the backbone of the polymer chain has been shown to be an effective way of increasing the adherence of the polymer to polar surfaces. The concentration of acid must be kept low to prevent association between the carboxyl groups which reduces surface adherence.<sup>53, 143</sup>



## B. Titanates

Increasingly of late, tetra esters of orthotitanic acid have been mentioned as coupling agents for glass, metals, and polymers. While no published mechanism for their action was found, it can be postulated that some of the ester linkages are hydrolyzed and coordination or condensation occurs between the resulting hydroxyl groups and surface groups. Furthermore, polymerization between adjacent titanate groups is likely, considering the effectiveness of poly(dialkyl titanates) as coupling agents.<sup>133</sup> Thus the interface between a polar surface and a titanate coupling agent might be pictured as:



The choice of alkoxy groups on the titanate determines the rate of hydrolysis, which has been reported to be the critical factor in proper surface preparation.<sup>112</sup> Butyl and isopropyl groups are the most widely used.

Alkyl titanates are also effective coupling agents for polyethylenes,<sup>112, 113, 150</sup> and interaction with such a non-polar surface is more difficult to envision. In any event, at least one alkoxy or hydroxy group is available for interaction with the adhesive resin by dipolar attraction or reaction. In addition to polyolefins, polyesters also benefit from titanate treatment prior to coating with polyethylene.<sup>37, 43</sup>



Aluminum, copper, and steel have been treated with a 'primer' composed of an amino silane mixed with tetraisopropyl titanate before bonding with a silicone adhesive.<sup>95</sup> Copper showed the greatest improvement in bond strength. A similar mixture of silane-titanate has been incorporated into polymers (vinyl, epoxy, or ester) for coating glass fibers.<sup>254</sup> Titanium chelates such as titanium acetyl acetonate, were also used in place of the tetra ester. Use of the chelates on glass surfaces has been investigated by equilibrium studies and the possibility has been raised of acidic surface sites catalyzing both the chelate hydrolysis and subsequent condensation with the surface silanol groups.<sup>344</sup>

### C. Amines

Amines have been used to a limited extent as coupling agents. Quaternary ammonium salts, which exhibit a positively charged nitrogen to bond with nucleophilic surface positions, have been used to increase adhesion of resins to glass.<sup>39</sup> A salt having at least one long-chain hydrocarbon radical is used to pretreat the glass surface.

Polyester films and fabrics show excellent adhesion to rubber if they are treated prior to vulcanization with a diamine such as 1,2-diaminoethane. Tear strength was increased forty fold over untreated polyesters.<sup>316</sup>

The effectiveness of certain amines in preventing aqueous corrosion of steel surfaces has promoted interest in their use as coupling agents. Amines have been shown to be chemisorbed in a mono-layer on the steel by virtue of the unbonded pair of electrons on the nitrogen and the vacant 'd' shell of iron.<sup>12, 97b, 115b</sup> Four-ethyl piperidine, which is particularly well adsorbed because of favorable polar and steric factors, bonds through the



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nitrogen atom and thus exposes the ethyl group for interaction with an adhesive resin. Further investigation is needed to determine optimum concentrations, substituent groups, substrates and adhesives to utilize piperidine coupling, although 4-aminomethylpiperidine has been shown to be beneficial in steel-epoxy systems.<sup>54b</sup>



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## VI. COMMERCIALLY AVAILABLE AGENTS

The following table is a compilation of commercially available coupling agents and their suppliers along with an elucidation of their structures. The list is divided along the same major lines as the entire paper, e.g., silanes, chromium complexes, and phosphorous compounds with appropriate subdivisions.

It should be noted that several of these compounds, particularly in the phosphorous section, are not marketed as coupling agents; however, since this review of the literature has found evidence that these types of compounds can be effective coupling agents, they are included in the compilation.

This collection is not meant to be complete with respect to commercial sources but presents a comprehensive list of types available.



TABLE I  
COMMERCIALY AVAILABLE COUPLING AGENTS

I. Silanes  $R-SiX_3$  ( $X$  = hydrolyzable group)

A. Saturated alkyl and haloalkyl  $R$  group

1. Methyltrichloro silane,  $CH_3-SiCl_3$  (General Electric S.C.-3100 and Stauffer SWS-442)
2. Ethyltriethoxy silane,  $CH_3-CH_2-Si(OEt)_3$  (General Electric SC-3735)
3.  $\gamma$ -Chloropropyltrimethoxy silane,  $Cl-CH_2-CH_2-CH_2-Si(OCH_3)_3$  (Dow Corning Z-8-0999)
4. Dimethyldichloro silane,  $(CH_3)_2-SiCl_2$  (Stauffer SWS-441)

B. Aryl and haloaryl  $R$  group

1. Phenyltrimethoxy silane,  $C_6H_5-Si(OCH_3)_3$  (Dow Corning 6071)
2.  $p$ -Chlorophenyltrichloro silane,  $Cl-C_6H_4-SiCl_3$  (K&K 13181)

C. Unsaturated alkyl  $R$  group

1. Vinyltriethoxy silane,  $CH_2=CH-Si(OEt)_3$  (Union Carbide A-151 and Dow Corning Z-6075)
2.  $\gamma$ -Methacryloxypropyltrimethoxy silane,  $CH_2=CH-\overset{\overset{O}{\parallel}}{C}-O-CH_2-CH_2-CH_2-Si(OCH_3)_3$  (Union Carbide A-174 and Dow Corning Z-6030)

D. Epoxy  $R$  groups

1.  $\gamma$ -Glycidoxypropyltrimethoxy silane,  $H_2C-\overset{\overset{O}{\parallel}}{C}-CH_2-O-CH_2-CH_2-CH_2-Si(OCH_3)_3$  (Dow Corning A-6040 and Union Carbide A-187)
2.  $\beta$ -(3,4-Epoxy cyclohexyl)-ethyltrimethoxy silane,  $O-\text{cyclohexane ring}-CH_2-CH_2-Si(OCH_3)_3$  (Union Carbide A-186)





E. Amino R group

1.  $\gamma$ -Aminopropyltriethoxy silane,  $\text{H}_2\text{N}-\text{CH}_2-\text{CH}_2-\text{CH}_2-\text{Si}(\text{OEt})_3$   
(Union Carbide A-1100)
2.  $\underline{\text{N}}\text{-}\beta\text{-(Aminoethyl)-}\gamma\text{-aminopropyltrimethoxy silane,}$   
 $\text{H}_2\text{N}-\text{CH}_2-\text{CH}_2-\text{NH}-\text{CH}_2-\text{CH}_2-\text{CH}_2-\text{Si}(\text{OCH}_3)_3$  (Union Carbide  
A-1120 and Dow Corning Z-6020)
3.  $\text{CH}_3-\text{O}-\overset{\text{O}}{\underset{\text{||}}{\text{C}}}-\text{CH}_2-\text{CH}_2-\text{NH}-\text{CH}_2-\text{CH}_2-\text{NH}-\text{CH}_2-\text{CH}_2-\text{CH}_2-\text{Si}(\text{OCH}_3)_3$   
(Dow Dorning C-600)

F. Mercapto R group

$\gamma$ -Mercaptopropyltrimethoxy silane,  $\text{HS}-\text{CH}_2-\text{CH}_2-\text{CH}_2-\text{Si}(\text{OMe})_3$   
(Union Carbide A-189)

II. Chromium Complexes (Acid Functions - Given)

- A. Methacrylo,  $\text{H}_2\text{C}=\text{C}(\text{CH}_3)-\text{CO}_2-$  (Dupont-Volan)
- B. Stearo,  $\text{CH}_3(\text{CH}_2)_{16}-\text{CO}_2-$  (Valchem--Valchrome 5015)

III. Tetra-alkyl Titanates ( $\text{R-O}$ )<sub>4</sub>Ti ( $\text{R-O}$  Groups Given)

- A. Isopropyl,  $(\text{CH}_3)_2\text{CH-O-}$  (Stauffer)
- B. n-Butyl,  $\text{CH}_3-(\text{CH}_2)_3-\text{O-}$  (Stauffer)
- C. 2-ethylhexyl,  $\text{CH}_3-(\text{CH}_2)_3-\text{CH}(\text{CH}_2-\text{CH}_3)-\text{CH}_2-\text{O-}$  (Stauffer)
- D. Cresyl,  $\text{CH}_3-\text{C}_6\text{H}_4-\text{O-}$  (Stauffer)

IV. Phosphorous Compounds (R Groups Given)

A. Phosphates

1. Monoalkyl acids  $\text{R-O}-\overset{\text{O}}{\underset{\text{||}}{\text{P}}}-(\text{OH})_2$ 
  - a. Butyl (Stauffer and Mobil)
  - b. Ethyl (Stauffer and Mobil)
  - c. Isoamyl (Stauffer and Mobil)



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B. Phosphonates  $R-\overset{\overset{O}{\parallel}}{P}-(OR)_2$

1. Diphenylphenyl (Stauffer)
2. Dimethylmethyl (Mobil)
3. Diethylethyl (Mobil)
4. Bis-(2-ethylhexyl)-2 ethylhexyl (Mobil)
5. Dibutylbutyl (Mobil)

C. Phosphites  $(R-O)_2P-OH$

1. Dimethyl (Stauffer)
2. Diethyl (Stauffer)
3. di-isopropyl (Stauffer)
4. Dioctyl (Stauffer)
5. Weslink E (Weston Chemical Co.) (exact structure unavailable)



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## VII. EVALUATION DATA

Most of the preceding discussion of the effectiveness of coupling agents in increasing dry and wet adhesive strength has been in qualitative terms. Some quantitative tests have been conducted but direct comparison between reported values is difficult because conditions were varied considerably. However, some of the reported data were selected for presentation in Table II to convey some insight into the adhesion-enhancing ability of the coupling agents. Items were chosen to represent as wide a variety of coupling agents, adhesives, substrates, and test conditions as possible from the available data. Bond improvement is reported as the ratio of bond strength with coupling agent to bond strength without coupling agent.

TABLE II

## QUANTITATIVE EVALUATION OF COUPLING AGENT EFFECTIVENESS

<u>Type</u>	<u>R-Group</u>	<u>Adhesive</u>	<u>Substrate</u>	<u>Test Type</u>	<u>Test Conditions</u>	<u>Bond Improvement (Treated/Untreated)</u>	<u>Reference</u>
<u>Silanes</u> <u>R-SiX<sub>3</sub></u>	$\alpha$ -amino propyl	poly olefin	glass-rubber	peel strength	dry	7.8	65
	$\gamma$ -amino propyl	epoxy-phenolic	glass laminate	tensile strength	dry	1.3	110
	vinyl	polyester	glass laminate	flex strength	2 hours boiling water	1.6	162
					dry	1.3	
	methyl methacrylate	polyester	glass laminate	flex strength	2 hours boiling water	3.0	162
					dry	1.4	
	3-glycidoxy propyl	polyester	glass laminate	adhesive strength	2 hours boiling water	4.2	228
					dry	1.4	
	3,4-epoxy cyclohexyl ethyl	polyester	glass laminate	adhesive strength	dry	1.2	228
					dry	1.4	
<u>Chromium Complex</u>	p-cyanophenyl	polybenzimidazole	glass laminate	flex strength	dry	1.4	240
	mercapto propyl	phenolic-acrylonitrile	aluminum	lap shear	2 hours boiling water	1.4	244
					73°F	2.1	
	amino propyl	phenolic-acrylonitrile	aluminum	lap shear	250°F	3.3	244
					73°F	1.6	
	3-glycidoxyl propyl	polyurethane	aluminum	lap shear	250°F	2.3	244
					75°F	1.7	304
					-300°F	1.3	304
					+200°F	4.4	304
	methacrylic acid	polyester	glass laminate	flex strength	dry	1.3	121
					3 hours boiling water	1.7	
<u>Carboxylic Acid</u> <u>R - CO<sub>2</sub>H</u>	trihydroxybenzoic	epoxy	glass laminate	flex strength	dry	1.2	121
					100 hours boiling water	2.2	
	salicylic	chloroprene-neoprene copolymer	canvas	peel strength	100°F	1.6	177
	succinimido-acetic	epoxy	aluminum	shear strength	200°F	1.7	274
					dry	1.6	
					48 hours boiling water	( $\frac{2000}{0}$ )	



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#### VIII. ACKNOWLEDGMENTS

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